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Explosion of a liquid film in contact with a pulse-heated solid surface detected by the probe-beam deflection method

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The threshold for explosive vaporization of a liquid layer on an opaque solid surface heated by an ultraviolet excimer pulsed laser is studied by a photoacoustic probe-beam deflection method. The probe beam traverses the liquid in the vicinity of the laser-heated liquid–solid interface. Below the explosion threshold, photoacoustic generation in the solid occurs only through a thermoelastic mechanism, which results mainly in shear waves that do not couple well into the liquid. Above the explosion threshold, photoacoustic pulses in the solid are also produced by explosive recoil, hence producing longitudinal pulses in the solid that couple well into the liquid after reflections. By setting the probe-beam refraction to detect longitudinal pulse echoes coupled back into the liquid, a sensitive detection of the explosive threshold can be established.

There is much recent interest in the study of the nucleation dynamics and explosive vaporization of a liquid film on a solid surface that is flash heated by a laser pulse.[1–3] This kind of liquid-film explosion is observed to lead to large forces exerted on microscopic particles on the surface, which leads to their rocketed ejection.[4–6] Previously, we have used a piezoelectric detector[7] as well as optical transmission[1,3] techniques to probe such nucleation dynamics. However, the piezoelectric probe is limited to a far distance ($\gtrsim$1 cm) from the irradiated source owing to its relatively large size; also, accurate detection of the transient photoacoustic (PA) pulse shape is difficult because of transducer ringing and the lack of bandwidth. The optical transmission probe is applicable only to certain thin samples with significant temperature-dependent transmissions. Here, we apply the PA probe-beam refraction techniques[8,9] to monitor for the first time to our knowledge multiple PA echoes generated in the solid sample both below and above the explosion threshold of the liquid at the interface. We show that a transition from a mainly shear-wave nature to a strongly longitudinal-wave nature for the PA wave in the solid signifies the onset of liquid explosion. This new technique is applicable to any opaque solids rather than only certain thin solids. Also, the probe beam can be set at a small distance ($\lesssim$1 mm) from the PA source so that excessive damping of the high-frequency signals can be avoided.

Figure 1 shows our experimental setup. We have built a compact probe-beam refraction sensing unit that contains a sample holder in the middle of a rigid structure. The probe He–Ne laser (633 nm) is mounted on one side of the structure, and a bicell position sensor is mounted on the opposite side. The rise time of the bicell is ~10 ns. The He–Ne beam is expanded by a fivefold beam expander to ~2.5 mm in diameter and is focused by a lens of focal length 5 cm. The theoretical beam waist (w) at the focal spot for a Gaussian beam is estimated to be ~16 μm. A direct measurement of w using a knife edge, a chopper, and the bicell gives a value for w of ~43 μm in air. The probe beam is ~0.4 mm from the surface of the sample. The KrF (248 nm, ~16 ns) UV excimer laser spot size is measured to be ~1 cm^2. A slit is used so that the width of the illuminated area of the sample is limited to 3 mm. This will optimize the probe refraction signal with the given beamwaist geometry.[10] By having the probe refraction measurement done in one compact and rigid unit, noise that is due to vibrations is eliminated to a large extent. In this experiment, we report only the PA refraction signal[9] and leave out the photothermal signal,[8] which could be recorded at a delay time of a few microseconds after the UV pulse. In addition, in the case of thin-film samples such as amorphous silicon (a-Si) deposited onto quartz, an additional probe based on optical transmission is measured simultaneously by using a cw diode (752-nm) laser.[1,3]

Various solid samples are studied. One solid sample is an a-Si thin film (0.2 μm thick) deposited onto fused quartz (1.5 mm thick). Previously, we

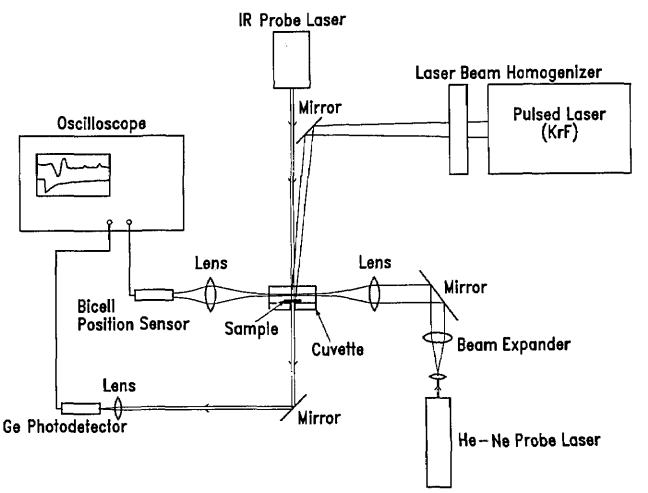

Fig. 1. Experimental scheme shown with simultaneous transmission and PA deflection probes.

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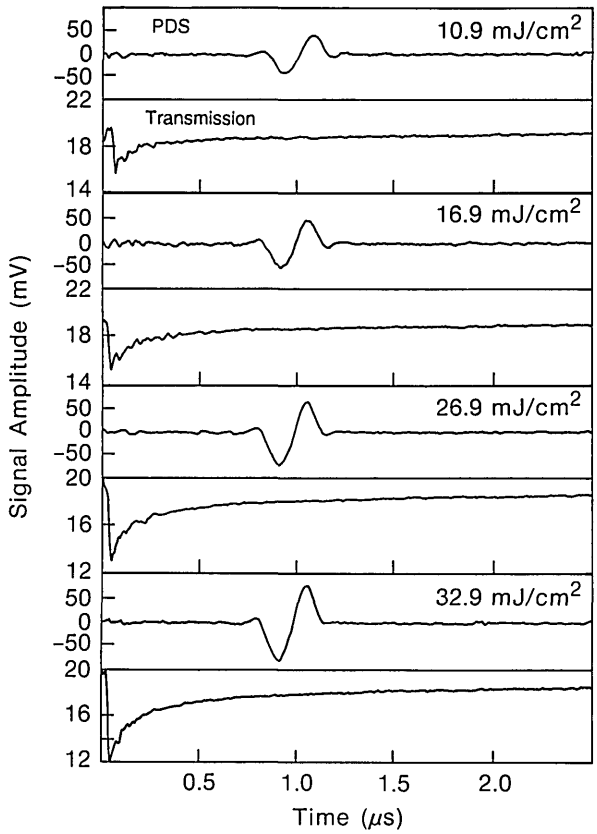

Fig. 2. Photoacoustic deflection signal (PDS) using a He–Ne laser (633-nm) and transmission (752-nm) signals for the a-Si sample in air with various incident KrF laser fluences.

have studied nucleation of liquids on such a substrate using an optical transmission technique.[1,3] Here, we first perform the measurements for the case of pulsed heating in air; the probe refraction is done simultaneously with the transmission signal, as shown in Fig. 2. There is no ablation of the substrate in the entire fluence range; the probe refraction signal or photoacoustic deflection signal in air remains constant in shape but just increases in amplitude linearly with the fluence. Also, the transmission drop at the end of the pulse gives the rise in surface temperature of the a-Si sample.[1,3] The signal is quite different when the a-Si sample is in contact with water under otherwise identical conditions, as shown in Fig. 3 for the range of KrF laser fluences used. The most striking feature is that now multiple echoes are detectable by the probe refraction signal, but only at higher fluence. The explanation of this striking effect is indicated in Fig. 4. At low fluence (i.e., below the liquid explosion threshold), thermoelastic expansion of the solid surface occurs, which generates mainly a shear PA pulse into the solid. The thermal coupling at the interface with the liquid leads to a thermal piston expansion of the liquid layer within the thermal diffusion length, but this only produces a weak longitudinal pulse into the solid owing to the weak acoustic coupling between the liquid and the solid because of acoustic impedance mismatch. The thermal piston effect in the liquid produces the first probe refraction pulse signal. The shear PA pulse propagates into the solid and is reflected at the sample back surface also mainly as a shear pulse with little mode conversion into longitudinal pulse at normal incidence. When this shear echo reaches the liquid–solid interface again, the shear wave cannot couple effectively into the liquid. This explains why there is at most weak echoes in the beam refraction signal below the liquid explosion

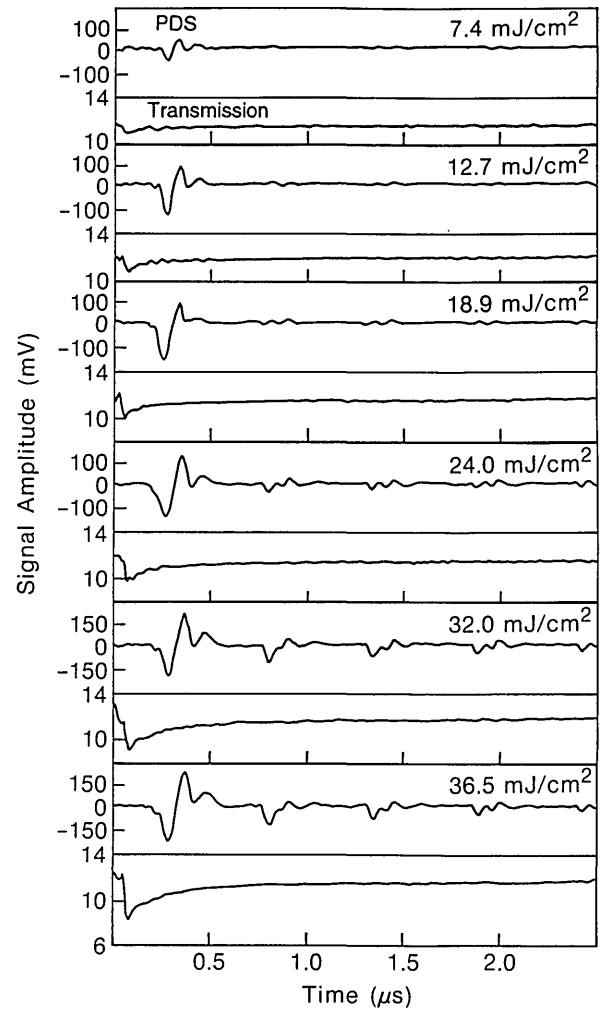

Fig. 3. Same as in Fig. 2 for the a-Si sample in H_2O. PDS, photoacoustic deflection signal.

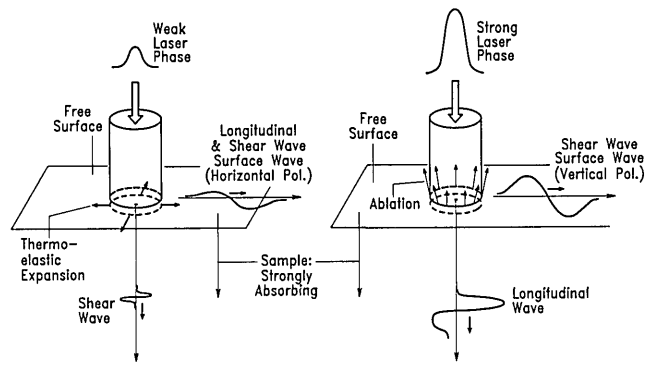

Fig. 4. Schematics of PA pulse generation that is due to (left) thermoelastic expansion at low fluence and (right) to explosion- or ablation-induced recoils for high fluence at a solid–fluid interface.

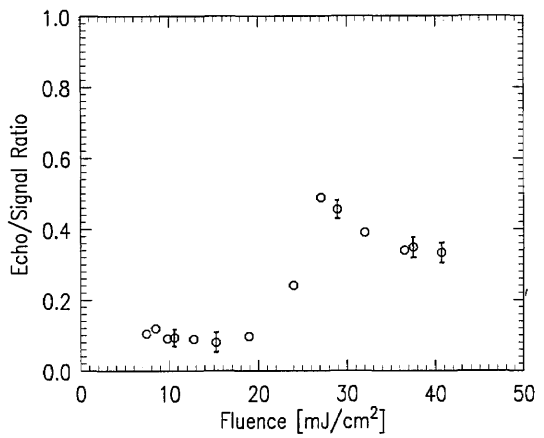

Fig. 5. Dependence of the ratio of the first echo to the initial pulse in the probe refraction signal for the a-Si–water system as a function of incident KrF laser fluence.

Table 1. Explosion Threshold Fluences (mJ/cm³) for Different Liquids on Different Substrates

	Liquids			
Substrates	H_2O	Isopropanol	Ethanol	Methanol
a-Si	24	11	10.5	10.5
a-C	34	18	17	16

threshold. However, above the explosion threshold, Fig. 4 indicates that a large recoil displacement at the solid–liquid interface is now possible, which causes a longitudinal PA pulse to be launched into the solid. This longitudinal pulse reflects back at the other sample surface as a longitudinal echo, which can be coupled effectively into the liquid to give a series of echoes in the probe refraction signal. We can get an idea of the efficiency of PA generation of the longitudinal component by dividing the amplitude of the first echo by the amplitude of the initial pulse in the probe refraction signal. This ratio for the a-Si–water case is shown in Fig. 5. We see that there is a sharp increase in this longitudinal echo efficiency within a narrow range of the laser fluence; the middle of range is taken as the fluence when significant longitudinal surface recoil at the liquid–solid interface occurs, i.e., liquid explosion occurs. With this definition as the threshold fluence for liquid explosion, we find that the explosion threshold for the a-Si–water interface is 24 mJ/cm². We have repeated such measurements for other liquids in contact with the same a-Si sample; these results are given in Table 1.

We have repeated the above measurements for an amorphous carbon (a-C) sample of thickness 0.8 mm. The probe refraction signals in air again increase linearly with fluence in the range studied with constant signal shape. There is no ablation of the sample at least up to 50 mJ/cm². The same a-C sample is then put into the cuvette with a liquid. Again, since there is no ablation of the sample within this range of fluence, any sudden increase in longitudinal echo amplitudes has to come from the liquid explosion. The liquid explosion threshold as defined above for the a-Si case for the a-C–water interface is found to be 34 mJ/cm², as given in Table 1 together with those of other liquids. We see two interesting features for the data summarized in Table 1. First, the explosion threshold for water is always much higher than that for an alcohol, because of the much higher transient temperature to which liquid water can be superheated before explosion.[1,3] Second, the a-C value is always ~50% higher than the a-Si value for the same liquid, indicating that the thermal effusivity of the a-C sample is ~50% larger than that of the a-Si sample (and this thermal effusivity of the solid sample dominates the temperature increase).

In conclusion, we have used for what is to our knowledge the first time a probe refraction technique as a sensitive monitor of near-threshold explosions of liquids in contact with a laser-heated solid surface. The signals indicate that the longitudinal PA pulse generation efficiency suddenly increases when liquid explosion occurs at the interface. This study is of use for the study of transient dynamics of superheated liquids as well as the mechanism of laser cleaning of surfaces.

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